(12) INTERNATIONAL APPLICATION PUBLISHED UNDER THE PATENT COOPERATION TREATY (PCT)

(19) World Intellectual Property Organization
International Bureau

(43) International Publication Date
8 February 2001 (08.02.2001)

PCT

(10) International Publication Number
WO 01/08795 A1

(51) International Patent Classification[7]: B01J 19/08

(21) International Application Number: PCT/KR00/00666

(22) International Filing Date: 26 June 2000 (26.06.2000)

(25) Filing Language: English

(26) Publication Language: English

(30) Priority Data:
1999/31713 2 August 1999 (02.08.1999) KR

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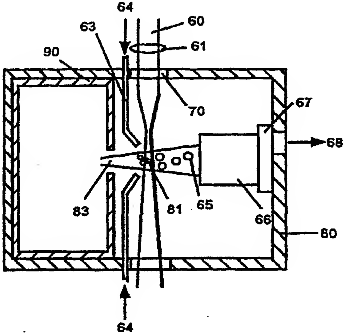
(81) Designated States *(national)*: DE, FI, IL, JP, US.

Published:
— *With international search report.*

(54) Title: FINE PARTICLE MANUFACTURING METHOD USING LASER BEAM



(57) **Abstract:** A method of manufacturing fine particles includes the steps of producing fine particles of a material targeted for manufacture by a physical or chemical method, irradiating a laser beam into the aggregates of the produced fine particles that exist in gases so that fine particles coalesce to form spherical particles, and trapping the spherical particles.

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FINE PARTICLE MANUFACTURING METHOD USING LASER BEAM

Technical Field

The present invention relates to a method for manufacturing fine particles including ceramic particles, metallic particles, glass particles or composite particles using a physical method or a chemical method, and more particularly, to a method for controlling the size, morphology, crystalline phase and composition of fine unagglomerate particles at high concentrations by facilitating coalescence of the fine aggregate particles by irradiating a laser beam into the produced fine aggregate particles.

Background Art

In order to physically manufacture fine particles, a solid or liquid material is heated to be evaporated, cooled to be super-saturated and is then recondensed. Here, for the purpose of evaporating the solid or liquid material, various heating methods are used, including heating using a laser beam, heating using an electrical heater or heating using an arc discharge. Once the solid or liquid material is evaporated, the evaporated material is super-saturated and recondensed in the same procedure to produce fine particles.

In a chemical method of manufacturing fine particles, the fine particles are produced by various chemical reactions induced by various means, such as plasma, microwave, an electrical heater or a combustion flame burner.

However, in the aforementioned physical and chemical method for manufacturing fine particles, in order to manufacture particles at high concentrations, aggregate particles shaped of a bunch of grapes and composed of small primary particles are generally produced. Since the aggregate particles have a collision cross-sectional area much larger than spherical particles of the same volume, they grow rapidly.

Now, a conventional method for manufacturing fine particles using a general physical method will be described with reference to FIG. 1.

FIG. 1 schematically shows a method for manufacturing particles by evaporating a material targeted for manufacture by an arc discharge. As shown, tungsten or graphite 1 is used as a positive electrode and a target material 2 is used as a negative electrode. Then, if a strong direct current (DC) is applied to the positive and negative electrodes, the target material 2 is evaporated within an evaporation space 5 to generate an arc 3.

Then, an inert gas 4 such as helium is injected in a direction indicated by an arrow to transfer the generated arc 3 to a particle trapping device 10. The transferred arc 3, that is, a vaporized material, passes through a cooler 9 to be recondensed and turned to be particles in a solid phase. The particles are trapped in the particle trapping device 10 by an electrical trapping method or a cold trapping method. Here, the pressures of the evaporation space 5, the cooler 9 and the particle trapping device 10 are controlled by using a vacuum pump connected through an exhaust duct 15.

However, according to the conventional method for manufacturing fine particles at high concentrations, a vaporized material is cooled in a cooler to cause super-saturation, thereby producing an aggregate 20 having a shape like that of a bunch of grapes, that is, many primary particles 21 are connected to one another, as shown in FIG. 2.

Now, a conventional chemical method for manufacturing fine particles will be described with reference to FIG. 5. As shown, a chemical precursor material 100 is injected into a chemical reactor 102 and the chemical precursor material 100 is heated using a heater 101 such as an electrical heater or a combustion flame burner, the heater 101 installed outside the chemical reactor 102. Alternatively, as shown in FIG. 6, a microwave 111 is formed in a reactor 102 using a plasma generator or microwave generator 110 installed outside the reactor 102, thereby causing a chemical reaction of the chemical precursor material 100.

In the case of producing oxide particles 120, high-purity oxygen, ozone or water vapor is contained in the chemical precursor material 100 injected into the reactor 102. In the case of producing non-oxide particles such as silicon (Si) particles, oxygen, ozone or water vapor is not injected.

If the chemical precursor material 100 injected into the reactor 102 is heated to a high temperature or becomes a plasma state, a chemical reaction occurs to nucleate extremely small particles 120 and the particles collide with each other to grow. Here, in view of economic manufacture, the particles must be in a high concentration per unit volume. To this end, a heavily concentrated chemical precursor material is injected into a reactor to produce aggregates 81 and 20, thereby accelerating the growth of particles.

In manufacturing thin films or bulk materials by sintering fine particles, aggregates cause flow and packing problems to become porous, which adversely affect the mechanical strength and optical and magnetic properties of the produced bulk materials.

Thus, in order to increase the quality and reliability in manufacturing various kinds of fine powders, it is important to form particles that have a uniform size distribution, have a spherical shape and are as small as possible in the nanometer scale, and also to control the crystalline phase of fine powders.

Disclosure of the Invention

To solve the above problems, it is an objective of the present invention to provide a method for manufacturing spherical fine particles smaller than those manufactured by the conventional method, by irradiating a laser beam into early stage fine aggregate particles formed by a conventional physical or chemical method to coalesce the aggregate particles so as to convert the same into spherical nanoparticles having the same volume as the fine aggregate particles, and by using the fact that the collision cross-sectional areas of the spherical nanoparticles are smaller than those of conventionally produced aggregate particles.

Since the nanoparticles coalesced by a laser beam collide with one another noticeably fewer times compared to the aggregate particles, the growth due to collision can be suppressed, thereby obtaining uniform, spherical particles which are finer.

It is another objective of the present invention to provide a method for manufacturing spherical fine particles having uniformly distributed compositions by

irradiating a laser beam into composite aggregate particles, thereby controlling the uniformity of composition in a particle. While fine particles are generated and grow in gases, the irradiation of laser beam on the aggregates causes rapid heating of particles and the transformed spherical particles may also experience rapid cooling right after passing the laser beam. This can be utilized to control the crystalline phase of fine particles..

To achieve the first objective of the present invention, there is provided a method of manufacturing fine particles includes the steps of producing fine particles of a material targeted for manufacture by a physical or chemical method, irradiating a laser beam into the aggregates of the produced fine particles so that fine aggregate particles coalesce to form spherical particles.

Also, the target material may consist of two or more components, and spherical composite particles are manufactured by irradiating a laser beam into the produced aggregates.

Therefore, according to the present invention, coalescence of particles is rapidly facilitated by irradiating high-power laser beam into aggregate particles at the time of formation thereof, thereby converting the aggregate particles into spherical particles. Also, high-concentration spherical particles smaller than conventionally produced particles can be manufactured by using the fact that spherical particles have smaller collision areas than the aggregate particles of the same volume.

In particular, the present invention is very advantageously used in manufacturing nanoparticles of 100 nm or less.

Brief Description of the Drawings

The above objectives and advantages of the present invention will become more apparent by describing in detail a preferred embodiment thereof with reference to the attached drawings in which:

FIG. 1 is a schematic diagram illustrating a conventional method for evaporating and condensing a material targeted for manufacture by an arc discharge;

FIG. 2 illustrates aggregates condensed by the method shown in FIG. 1;

FIG. 3 is a schematic diagram for explaining a method for evaporating a

material targeted for manufacture by irradiating a laser beam;

FIG. 4 is a schematic diagram for explaining a method for evaporating a material targeted for manufacture by an electrical resistance heating method;

FIG. 5 is a schematic diagram for explaining a method for manufacturing fine particles by inducing a chemical reaction of reactants using a conventional heater;

FIG. 6 is a schematic diagram for explaining a method for manufacturing fine particles by inducing a chemical reaction of reactants using a conventional plasma or microwave generator;

FIG. 7 is a schematic diagram illustrating an apparatus for adopting the method for manufacturing fine particles according to an embodiment of the present invention;

FIG. 8 is a schematic diagram partially illustrating an apparatus for adopting the method for manufacturing fine particles according to another embodiment of the present invention; and

FIG. 9 is a schematic diagram partially illustrating an apparatus for adopting the method for manufacturing fine particles according to a still another embodiment of the present invention.

Best mode for carrying out the Invention

Preferred embodiments of the present invention will now be described in more detail with reference to the accompanying drawings.

FIG. 7 is a schematic diagram illustrating an apparatus for adopting the method for manufacturing fine particles according to an embodiment of the present invention. As shown, the fine particle manufacturing apparatus includes an evaporator 90 installed in a housing 80, for evaporating a material targeted for manufacture, a gas nozzle 63 for injecting gas into the evaporated material to be condensed, a beam irradiation apparatus (not shown) for irradiating a laser beam 60 into fine aggregate particles produced from the evaporated material, and a cooler 66 and a trapper 67, both for trapping the produced particles.

Reference numeral 61 denotes a focusing lens for focusing a laser beam, reference numeral 70 denotes a window for projecting the laser beam, and reference

numeral 68 denotes an exhaust path connected to a vacuum pump for adjusting the pressures of an evaporation space and a trapping space.

The evaporator 90 can be any conventional evaporation apparatus that is used in physical deposition. In other words, an evaporation method using an arc discharge, disclosed in U.S. Patent Nos. 4,732,369 and 5,472,749, an evaporation method using laser beam or electron beam irradiation disclosed in U.S. Patent No. 5,534,314, or an evaporation method using electrical resistance or inductive heating disclosed in U.S. Patent No. 5,618,475, can be used, which will now be described in detail.

The apparatus and method for evaporating a material targeted for manufacture has been described above with reference to FIG. 1, and a further explanation thereof will not be given. FIG. 3 shows an apparatus based on evaporation using laser beam irradiation.

Referring to FIG. 3, a laser beam 30 is irradiated through a lens 31 and a window 32 into a solid or liquid material 22 targeted for manufacture, prepared within an evaporation space 25 of a chamber 34 to evaporate the target material 22, so that it becomes a vapor phase 23. The vapor-phase material is carried by an inert gas 4 sprayed by a separate supply means so that it is exhausted through a vapor spray orifice 33.

The evaporation apparatus using an electron beam is substantially the same as the evaporation apparatus using laser beam irradiation, except that a focusing lens and a window corresponding to the electron beam are provided.

An alternative evaporation apparatus based on electrical resistance heating is shown in FIG. 4. As shown, a rod-shaped target material 42 is placed into an evaporation space 35 of a chamber 46 and a DC or AC electrical field is applied from a power source 40 to an electric resistor 44 surrounding the evaporation space 35, for heating. A ceramic insulator 45 is provided around the electric resistor 44 to reduce heat loss.

One end 42′ of the target material 42 is melted by heating the resistor 44 and a vapor-phase material evaporated therefrom is sprayed by a gas 4 through a vapor spraying orifice 43. Alternatively, inductive heating may be used instead of

electrical resistance heating.

Now, a method for manufacturing fine particles using the aforementioned apparatus according to the present invention will be described in detail.

Referring back to FIG. 7, a material targeted for manufacture (not shown) is evaporated by the aforementioned evaporation apparatus. The target material which can be adopted in the present invention is not specifically limited, inclusive of ceramic, metal, carbon, glass or composite, and compounds produced by combination thereof, can be used.

Then, a gas is supplied to inject a vapor-phase jet 83. The supplied gas is preferably an inert gas such as helium. Also, according to the kind of particles produced, a reactant gas such as oxygen, nitrogen or argon can be used, not being limited by this embodiment of the present invention.

The vapor-phase jet 83 is exhausted through a vapor exhausting orifice (not shown). Gas 64 is additionally injected into the exhausted vapor-phase jet 83 through a gas nozzle 63 spaced an appropriate distance apart from the vapor exhausting orifice, thereby recondensing the jet 83 to produce particles. As described above, the produced particles include small-sized aggregate particles 81 when high-concentration particles are produced. Although not shown, the particles may be produced by expansion as well as additional gas injection.

According to the present invention, a laser beam 60 is irradiated into the produced aggregate particles that exist in gases through a focusing lens 61. The small-sized aggregate particles 81 are coalesced by laser beam irradiation, so that they form spherical nanoparticles 65. The produced spherical particles have a collision cross sections smaller than those of aggregates. Thus, the growth rate of the spherical particles is reduced and small-sized spherical particles can be obtained by trapping the same using the conventional cooler 66 and trapper 67.

According to another aspect of the present invention, as shown in FIG. 8, in order to increase the irradiation efficiency of a laser beam. a plurality of reflecting mirrors 69a and 69b are installed. The laser beam travels in a plurality of paths through the reflecting mirrors 69a and 69b. thereby further increasing the coalescence effect of particles. Also, although not shown. a cylindrical lens is

installed, instead of the focusing lens 61 shown in FIG. 7, and a plane laser beam having passed through the cylindrical lens is reflected by the reflecting mirrors 69a and 69b, thereby coalescing the particles.

The present invention can also be applied to a chemical process in addition to the above-described physical process, which will now be described with reference to FIG. 9.

Referring to FIG. 9, a fine particle manufacturing apparatus according to the present invention includes a chemical precursor material injector (not shown) for injecting a reactant 100 into a chemical reactor 102, a reaction inducing apparatus 115 for chemically reacting the injected reactant 100 to produce particles, a beam irradiation apparatus (not shown) for irradiating a laser beam 160 into small-sized aggregate particles 81 produced in a chemical reactor 102, and a cooler 166 and a trapper 167, for trapping the produced particles. Here, reference numeral 168 denotes an exhaust path connected to a vacuum pump, for adjusting the pressures of a reactor space and a trapping space. Reference numeral 120 denotes nucleated particles initially produced by the chemical reaction of the reactant, and reference numeral 65 is small-sized spherical particles converted from the small-sized aggregate particles 81 and coalesced by a laser beam 60

The reaction inducing apparatus 115 can be any conventional apparatuses that chemically produce particles. In other words, a heating apparatus (see FIG. 5) for inducing a chemical reaction by heating a reactant, or a plasma or microwave generator (see FIG. 6) for inducing a chemical reaction by generating plasma or microwave inside a chemical reactor, can also be used in the present invention.

Now, a method for manufacturing fine particles using the aforementioned apparatus will be described.

The reactant 100 is injected into the chemical reactor 102 and then the reactant 100 is subjected to a chemical reaction by the reaction inducing apparatus 115 installed outside the chemical reactor 102 to nucleate particles 120. The nucleated particles 120 are turned into the small-sized aggregate particles 81 by collision.

Then, the laser beam 160 which is focused through the focusing lens 161 and

the window 170 is irradiated into the aggregate particles 81 so that the aggregate particles 81 coalesce, thereby producing spherical particles 65 while having the same volume. The thus-produced spherical particles 65 have collision cross-sectional areas much smaller than the aggregate particles 81, thereby retarding the growth of particles and finally obtaining much smaller spherical nanoparticles.

In the case of producing oxide particles, a chemical precursor material containing high-purity oxygen, ozone or water vapor is injected into the reactor. In the case of producing non-oxide particles, oxygen, ozone or water vapor is not injected.

.In the case where a material targeted for manufacture is a composite consisting of multiple components. a laser beam is irradiated into small-sized composite particles manufactured by the aforementioned manufacturing apparatus to facilitate coalescence of particles, thereby obtaining spherical composite particles.

According to the method for manufacturing small-sized spherical particles by enhancing the coalescence of particles that exist in gases using laser beam heating, spherical particles smaller than the conventional particles can be manufactured by irradiating a laser beam into a region at which small aggregate particles are formed in the conventional physical or chemical process. Also, the present invention can also be applied in manufacturing composite nanoparticles.

Although the invention has been described using an exemplary preferred embodiment, various modifications and changes can be effected by one skilled in the art within the scope of the present invention, and it is intended the scope of the invention be defined by the claims appended thereto.

Industrial Applicability

The present invention can be applied to manufacture of fine particles including ceramic particles, metallic particles, glass particles or composite particles using a physical or chemical method.

What is claimed is:

1. A method for manufacturing fine particles, comprising the steps of:

producing fine particles of a material targeted for manufacture by a physical or chemical method;

irradiating a laser beam into the aggregates of the produced fine particles that exist in gases so that fine particles coalesce to form spherical particles and the growth of particles is retarded; and

trapping the spherical particles.

2. The method according to claim 1, wherein the target material consists of two or more components, and spherical composite particles are manufactured by irradiating a laser beam into the produced aggregates.

3. The method according to claim 1, wherein the irradiation of laser beam on the aggregates causes particles to be rapidly heated and the transformed spherical particles are rapidly cooled right after passing the laser beam, thereby controlling the crystalline phase of fine particles.

FIG. 1

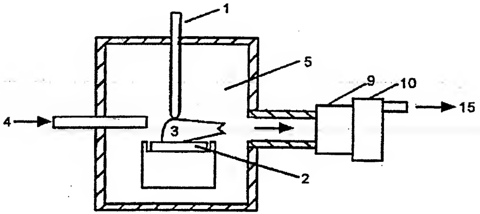

FIG. 2

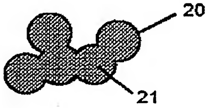

FIG. 3

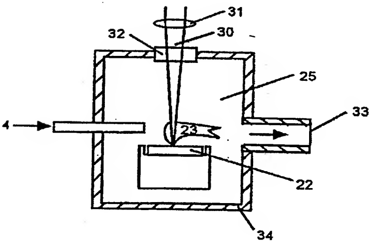

FIG. 4

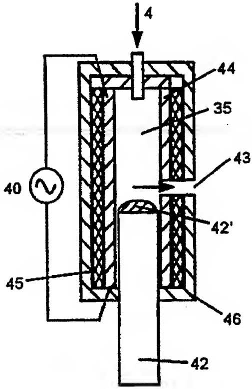

FIG. 5

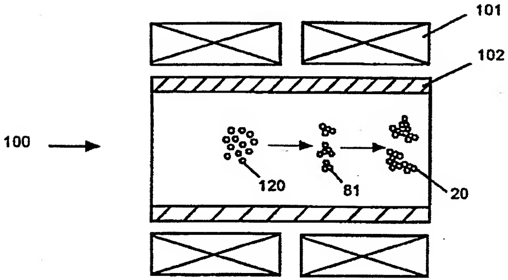

FIG. 6

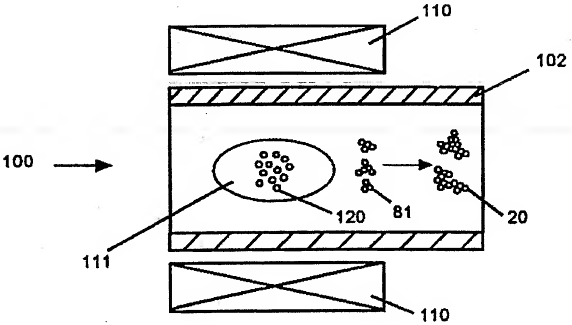
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FIG. 7

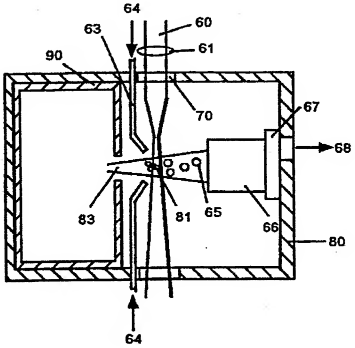
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FIG. 8

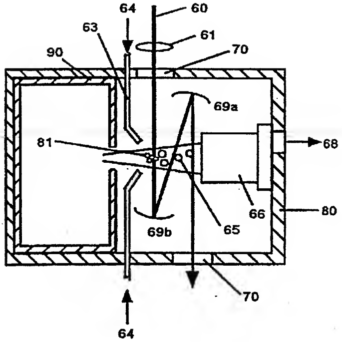
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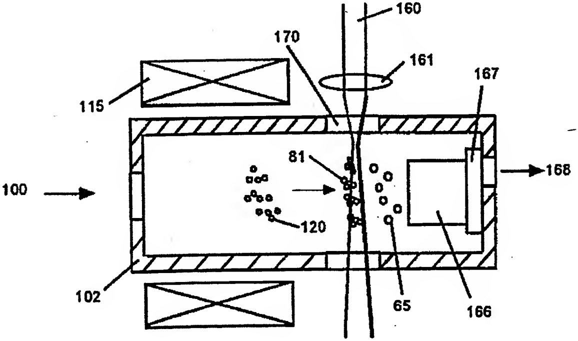
FIG. 9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INTERNATIONAL SEARCH REPORT

International application No.
PCT/KR00/00666

A. CLASSIFICATION OF SUBJECT MATTER

IPC7 B01J 19/08

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimun documentation searched (classification system followed by classification symbols)

IPC7 B01J 19/08, B22F 9/02

Documentation searched other than minimun documentation to the extent that such documents are included in the fileds searched

Korean Patents and applications for inventions since 1975

Electronic data base consulted during the intertnational search (name of data base and, where practicable, search trerms used)

NPS, PAJ

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
Y	JP 63-33507 A (MITSUBISHI METAL CORP) 13 FEBRUARY 1988 see the whole document	1-3
Y	JP 60-51539 A (TOYOTA CENTRAL RES & DEV LAB INC) 23 MARCH 1985 see the whole document	1-3
Y	JP 5-261265 A (NIPPION SHEET GLASS CO LTD) 12 OCTOBER 1993 see the whole document	1-3
A	EP 379360 A2 (IDEMITSU KOSAN CO) 25 JULY 1990 see the whole document	1-3
A	JP 1-242143 A (AGENCY OF IND SCIENCE & TECHNOL) 27 SEPTEMBER 1989 see the whole document	1-3

☐ Further documents are listed in the continuation of Box C.

☒ See patent family annex.

* Special categories of cited documents:

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"P" document published prior to the international filing date but later than the priority date claimed

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"&" document member of the same patent family

Date of the actual completion of the international search	Date of mailing of the international search report
10 OCTOBER 2000 (10.10.2000)	11 OCTOBER 2000 (11.10.2000)

Name and mailing address of the ISA/KR	Authorized officer
Korean Industrial Property Office Government Complex-Taejon, Dunsan-dong, So-ku, Taejon Metropolitan City 302-701, Republic of Korea	CHOI, In Sun
Facsimile No. 82-42-472-7140	Telephone No. 82-42-481-5567

INTERNATIONAL SEARCH REPORT

Information on patent family members

International application No.
PCT/KR00/00666

Patent document cited in search report	Publication date	Patent family member(s)	Publication date
JP 63-33507 A	13.02.88	none	
JP 60-51539 A	23.03.85	none	
JP 5-261267 A	12.10.93	none	
EP 379360 A2	25.07.90	US 5064517 A	12.11.91
		US 5207878 A	04.05.93
		JP 2-190408 A2	26.07.90
		JP 2-188410 A2	24.07.90
		JP 3-16901 A2	24.01.91
JP 1-242143 A	27.09.89	none	